FINAL REPORT*

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I-129 DOSIMETRY:

A DEMONSTRATION PROJECT FOR THE USE OF ^{129}I AS A SURROGATE FOR ^{131}I IN BELARUS AND OTHER REGIONS CONTAMINATED BY CHERNOBYL FALLOUT

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INTRODUCTION

Several hundred thousand children's thyroids were measured with survey meters within weeks after the Chernobyl accident to reconstruct thyroid doses in regions of Belarus, Ukraine, and Russia contaminated by radionuclides. For areas where such measurements were either absent or inadequately performed, it was hoped that the deposition density of ^{137}Cs might be a useful surrogate for the deposition density of ^{131}I and that this information, coupled with the use of ecological models, could be used to infer doses to the thyroid. However, we now know that the ratios of ^{131}I-to-^{137}Cs vary by factors of more than 300 over the country of Belarus and thus this ratio is unreliable for thyroid dosimetry (e.g., Straume et al. 1996, and data presented here).

A better surrogate for the retrospective determination of the deposition density of ^{131}I would be ^{129}I, which has a half life of 1.57×10^7 years and which has been reported to have a mean-residence time in the first meter of surface soil of approximately 1000 years (Kocher 1991). Due to its low specific activity and its lack of emission of a high-energy gamma ray, ^{129}I is not measured with sufficient sensitivity by conventional counting techniques, but it can be measured with techniques such as neutron-activation analysis and accelerator-mass spectrometry (AMS). For soil samples, the detection limit for ^{129}I measured by neutron activation appears to be in the range of 1 to 100 μBq sample^{-1} (Katagiri et al. 1990; Muramatsu et al. 1990; Doshi et al. 1991); this activity range is equivalent to ~10^9 to ~10^{11} ^{129}I atoms per sample. A similar detection limit has been reported for thyroids (Handl et al. 1990). In contrast, the detection limit for ^{129}I measured by AMS is ~10^6 atoms per sample (Elmore and Phillips 1987), orders of

magnitude lower than the detection limits cited above for neutron activation. Importantly, the increased sensitivity of AMS is in fact necessary for the measurement of depth profiles in soil of sufficient precision to reconstruct ^{129}I deposition densities in Belarus, and by inference, in the Ukraine and Russia as well (Straume et al. 1996 and 1997).

In 1993, a feasibility study was initiated to determine whether the deposition of ^{129}I released from Chernobyl could be measured reliably using soil sampling and extraction methods to be developed as part of the feasibility study and using AMS for isotopic analysis. Twenty-two sites in 11 settlements in Belarus were sampled by coring to 30 cm depths in soil. Efforts were taken to ensure that the sampling sites had not been disturbed since the Chernobyl accident (which, in retrospect, turned out to be correct most of the time). Results from the feasibility study have been published (Straume et al. 1996) and demonstrated that our AMS-based approach, together with iodine extraction methods developed for that project (Marchetti et al. 1994 and 1997), can be used reliably to measure ^{129}I in soil samples from background levels to the highest concentrations in the 30-km zone. The feasibility study also confirmed previous findings by others that the ratio of radioiodine-to-radiocesium varied substantially over space and may therefore not be reliable as a general basis for thyroid dosimetry.

In 1996, a follow-on study was initiated with the objectives to use the methods developed in the feasibility study to measure ^{129}I, total iodine, and ^{137}Cs in soil samples from settlements throughout the country of Belarus. To accomplish this, a soil-sampling expedition was undertaken in 1997.

MATERIALS AND METHODS

Soil sampling

Soil-sampling expeditions were conducted in Belarus in May 1993 and June 1997 and included scientists from the USA, Belarus, Russia, and Germany. The 1993 sampling expedition has been described previously (Straume et al. 1996). Settlements and sampling sites for both expeditions are listed in Table 1 and plotted on the map of Belarus in Figure 1.

For the 1997 sampling expedition, the settlements were selected so that soil would be sampled from all major regions of Belarus. The specific sampling sites within each settlement were selected based on (1) information from local officials that the sites had not been disturbed since the accident, (2) our measurements of ^{137}Cs-photon flux as determined by field-gamma spectrometry and of external gamma-exposure rates, and (3) information indicating that the sites were representative of the settlement. As far as possible, sites were selected in fields away from buildings or trees that might have influenced local deposition patterns. At each sampling site, five cylindrical cores of 5-cm diameter and 30-cm depth were obtained in a 5 m x 5 m square and one in the center. Generally, each core was divided into 2 depth layers (0-15 cm and 15-30 cm) and the five cores combined for each depth layer. For two settlements that were sampled in 1993 and resampled in 1997 (Holochie and Veprin), full depth profiles were obtained for intercomparison, i.e., 0-2.5, 2.5-5, 5-10, 10-15, 15-20, 20-25, and 25-30 cm, and the layers from the same depths combined for each site. The approach of obtaining multiple cores in a sampling site should average possible differences in radionuclide concentrations within the sampling site.

Table 1. Soil sampling sites in Belarus

Settlement & Sampling site	Map #	Settlement & Sampling site	Map #
Tulgovitchi-1 (93)	1	Grandichi-1 (97)	39
Tulgovitchi-2 (93)	2	Grandichi-2 (97)	40
Deriazshna-1 (93)	3	Lunno-1 (97)	41
Deryashnya-2 (93)	4	Lunno-2 (97)	42
Deryashnya-3 (93)	5	Pogonnoe-1 (93)	43
Veprin-1 (93)	6	Pogonnoe-2 (93)	44
Veprin-2 (93)	7	Izabelino-1 (97)	45
Gorodische-1 (97)	8	Izabelino-2 (97)	46
Gorodische-2 (97)	9	Shevernichi-1 (97)	47
Boloto-1 (97)	10	Shevernichi-2 (97)	48
Boloto-2 (97)	11	Osipovichi-1 (97)	49
Pruzhany-1 (97)	12	Osipovichi-2 (97)	50
Pruzhany-2 (97)	13	N. Borschevka-1 (93)	51
Ostrogliady-1 (93)	14	N. Borschevka-2 (93)	52
Ostrogliady-2 (93)	15	Chudiany-1 (93)	53
Bogino-1 (97)	16	Rotki-1 (97)	54
Bogino-2 (97)	17	Bobr-1 (97)	55
Obrub-1 (97)	18	Bobr-2 (97)	56
Obrub-2 (97)	19	Pleschenitsy-1 (97)	57
Holochie-1 (93)	20	Pleschenitsy-2 (97)	58
Holochie-2 (93)	21	Losha-1 (97)	59
Yankovichi-1 (97)	22	Losha-2 (97)	60
Yankovichi-2 (97)	23	Zabolotye-1 (97)	61
Lyntupy-1 (97)	24	Zabolotye-2 (97)	62
Lyntupy-2 (97)	25	Krynka-1 (97)	63
Babinichi-1 (97)	26	Krynka-2 (97)	64
Babinichi-2 (97)	27	Kulikovka-1 (97)	65
Korma-2 (97)	28	Kulikovka-2 (97)	66
Korma-1 (97)	29	Bokhany-1 (97)	67
Stodolichi-1 (97)	30	Bokhany-2 (97)	68
Stodolichi-2 (97)	31	Volkovichi-1 (97)	69
Simonichi-1 (97)	32	Volkovichi-2 (97)	70
Simonichi-2 (97)	33	Veprin-1(97)	71
Yanovka-1 (93)	34	Veprin-2(97)	72
Yanovka-2 (93)	35	Bentyai-1 (93)	73
Porechye-1 (97)	36	Bentyai-2 (93)	74
Porechye-2 (97)	37	Ostrovskie-1 (93)	75
Holochie-1 (97)	38	Ostrovskie-2 (93)	76

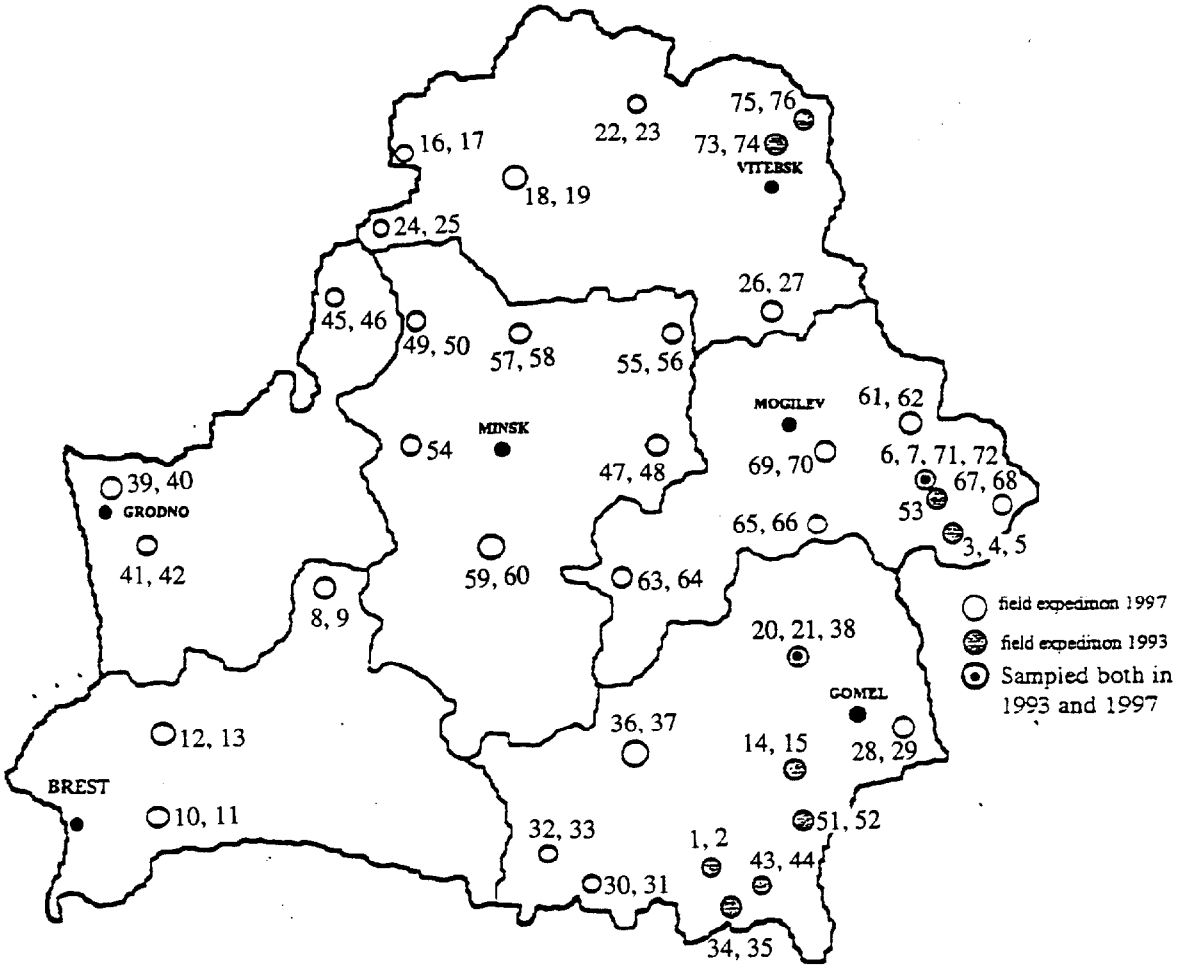

Figure 1. A map of Belarus with sampling locations. Sampling expeditions in 1993 (•) and 1997 (O) are included. Also included are the locations sampled in both 1993 and 1997 (⊙).

Soil cores were processed at the Research and Clinical Institute of Radiation Medicine and Endocrinology (RCIRME) in Minsk, Belarus. Processing involved drying at room temperature and weighing daily until no further weight loss was detected. Each soil sample was then mixed and sieved through a 3-mm sieve. Aliquots of the homogenized soil samples were provided to the University of Utah for iodine extraction, measurement of ^{129}I, and total iodine, and to the RCIRME for measurement of ^{137}Cs by gamma spectrometry.

Extraction and quantification of total iodine in soil

Aliquots of the processed soil samples (typically, one to three grams) were weighed in a porcelain-combustion boat and placed in a quartz-tube furnace according to the method of Marchetti et al. 1997. One end of the quartz tube was connected to a supply of oxygen and the other end was connected to a bottle containing a solution to trap iodine. Between the combustion boat and the trap there was a quartz-wool plug preheated to 1000°C by an auxiliary furnace. The trapping solution was prepared by adding 0.5 g of Na_2SO_3 to 75 mL of 0.1 M KOH. Once the combustion boat was placed at the center of the main furnace, the oxygen flow was adjusted to 50–100 mL min^{-1} and the temperature of the furnace was set at 1000°C. The furnace required about 20 min to reach that temperature. Once the furnace was at 1000°C, combustion was allowed to proceed for 90 min. Under these conditions, essentially all iodine present in the sample was volatilized and carried in the oxygen stream, ultimately as molecular iodine. Any other volatile compounds of iodine that may have been released as the furnace heated up would have been combusted at the preheated quartz plug. The iodine then reacted with the solution to form iodide that remained in the liquid trap.

After combustion, the trap solution was transferred to a 100-mL volumetric flask and diluted to the mark with deionized water. A 50-mL aliquot of the solution was placed in a 250-mL beaker followed by the addition of 5 to 20 mL of a 1000-ppm

solution of iodide as carrier prepared from KI and 1 mL of concentrated HNO_3. Finally, a solution of 5% $AgNO_3$ was added in excess to precipitate AgI. The precipitate was left overnight to settle, transferred to a centrifuge tube, washed with deionized water three times, and dried in an oven at 70°C. The dried precipitate was thoroughly mixed with an equal volume of silver powder. About 5 mg of this mix was loaded in a sample holder for the AMS measurement of ^{129}I. The remaining 50 mL of solution were separated into two 25-mL aliquots. A determination of total iodine was performed on each aliquot using a gas chromatograph (GC) with an electron-capture detector. The detection limit of this method has been demonstrated to be <0.05 μg iodine per g soil (Marchetti et al. 1994, 1997), well below the concentration range of ~1 μg/g commonly observed in soils.

^{129}I measurements

Measurements of ^{129}I were performed using the AMS facility at Purdue University (PrimeLab). Isotopic measurements by AMS are relative measurements, i.e., they measure ratios of counts that are scaled to $^{129}I/I$ using a NIST standard. To obtain the actual number of ^{129}I atoms, the measured ^{129}I-to-I ratio must be multiplied by the total number of iodine atoms in the sample. For the present case, the total number of iodine atoms in the sample is the number of iodine atoms in the soil sample (from the GC measurement) plus the number of iodine atoms in the KI carrier (known from the amount of carrier added). Note that essentially all of the iodine in the AMS sample is in fact from the carrier, i.e., typically 10 mg carrier compared with μg quantities in the soil sample itself.

^{137}Cs measurements

About 500 g of soil from each soil layer were analyzed using a gamma-ray spectrometer with a GeLi detector. All gamma-ray-emitting nuclides with activities

above the limit of detection were recorded. Here, only the ^{137}Cs results are presented for comparison with the ^{129}I results; measured activities of ^{137}Cs were converted to atoms for intercomparison.

Measurements of ^{137}Cs were made in Minsk with QA measurements at LLNL and the University of Utah. The QA measurements have demonstrated good agreement between the labs.

Deposition densities

The densities of ground deposition for ^{129}I and ^{137}Cs were obtained from the soil core measurements. For ^{129}I, the deposition density at each sampling site was obtained from the measured number of atoms of ^{129}I per gram of soil minus background times the total number of grams of soil sampled. This deposition density was then scaled to a ground surface area of 1 m^2. For ^{137}Cs, the same procedure was used with the exception that the gamma-ray spectrometry measurement results in Bq per gram soil were first converted to atoms per gram.

The deposition densities for ^{131}I at the time of the accident were inferred from the measured ^{129}I densities by dividing atoms of ^{129}I per m^2 by 15. The proportionality constant of 15 was obtained from measurements of archived soil samples, estimates based on reactor fuel burn-up, and $^{129}I/^{131}I$ ratios from measurements of rain water shortly after the accident (Straume et al. 1996). One archived soil sample was obtained that was measured for ^{131}I soon (11 May 1986) after the accident in 1986 and then re-measured for ^{129}I in 1994. This provided a direct measure of the ratio of ^{129}I-to-^{131}I in the soil sample, which was collected on 1 May 1986, from Holochie, a settlement north of Gomel. Four aliquots from this sample were measured for ^{129}I in 1994. The results were 1.18×10^{10}, 1.49×10^{10}, 1.20×10^{10}, and 1.04×10^{10} atoms of ^{129}I per g of soil. When compared with the decay-corrected results for ^{131}I of 1.0×10^{9} atoms per g (for 26 April 1986), the atomic ratios for ^{129}I-to-^{131}I are 11, 14, 12, and 10, respectively. These values

result in a mean of 12 and a standard deviation of 3. Estimates based on fuel burnup resulted in activity ratios ranging from 1.5×10^{-8} to 2.1×10^{-8} (Ermilov et al. 1993), which convert to atomic ratios of 11 to 15. Also, measurements made on three rainwater samples collected in Israel and Germany within a couple of weeks of the accident resulted in an unweighted mean of 21±13 and a weighted mean of 15±3.

Taken together, results indicate that the atomic ratio of ^{129}I-to-^{131}I released from the Chernobyl reactor during the 1986 accident was in the 10 to 20 range.

RESULTS

Iodine extraction

The efficiency of our iodine-extraction method was determined using standard-reference materials, including NIST soils (Marchetti et al. 1997). In brief, the recovery of total iodine from the reference soil samples ranged from 89.0% to 97.7%. It is expected that the recovery efficiency would be even higher (essentially 100%) for iodine deposited from Chernobyl because such deposition would mostly adhere to the surfaces of minerals, whereas much of the iodine in the NIST soils was actually trapped inside the crystalline structure of the minerals.

Comparison of soil-depth profiles, 1993 vs. 1997

Sites in two settlements in Belarus were sampled in both 1993 and 1997. The settlements resampled were Veprin and Holochie.

Both the ^{129}I and ^{137}Cs depth profiles obtained from the 1997 core samples agree very well with those obtained four years previously in 1993. The ^{129}I profiles obtained for Veprin-2 are seen in Fig. 2, and the ^{137}Cs profiles are seen in Fig. 3.

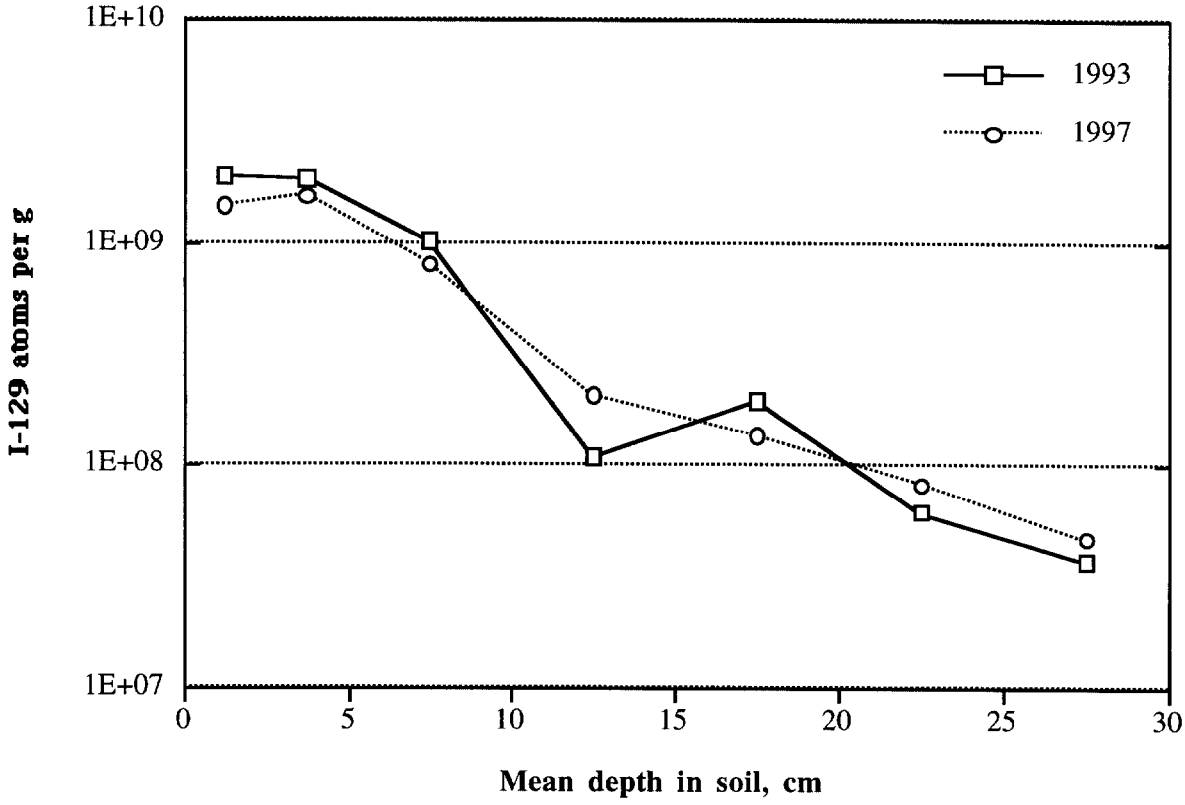

Figure 2. Depth profiles of ^{129}I measured in soil cores collected in 1993 and 1997 from Veprin, Belarus.

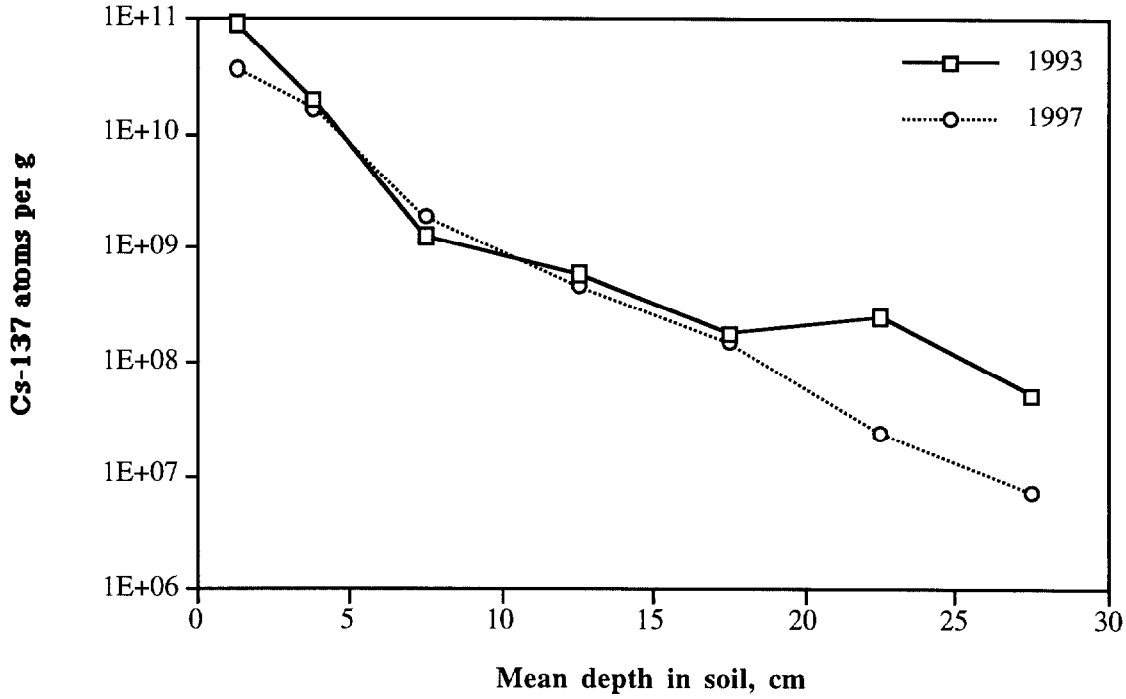

Figure 3. Depth profiles of ^{137}Cs measured in soil cores collected in 1993 and 1997 from Veprin, Belarus. These measurements were made in the same core samples subsequently used to measure ^{129}I and seen in Fig. 2.

Deposition of ^{129}I

Results for the deposition densities of ^{129}I are listed in Table 2. Again, these sampling sites correspond to the sampling locations indicated on Figure 1.

The data show a range of deposition densities spanning more than two orders of magnitude from essentially background in Ostrovskie (Vitebsk Oblast) to a maximum in Pogonnoe within the 30 km zone.

The data also show that although there are large regional differences (as expected) in the deposition densities for ^{129}I, the intra-settlement differences tend to be only a factor of about two. That is, when comparing the deposition densities obtained from the different sampling sites within a settlement, the mean difference is a factor of 1.95±1.10.

If the same sampling site, but different locations within the site, is re-sampled the deposition densities obtained from the two samplings are even closer. This is illustrated in Figure 2 where the samplings were done four years apart resulting in deposition densities of 2.07×10^{14} atoms ^{129}I per m^2 in 1993 and 1.41×10^{14} in 1997.

Deposition of ^{137}Cs

Results for the deposition densities of ^{137}Cs are listed in Table 3. Again, these sampling sites correspond to the sampling locations indicated on Figure 1.

The data show a range of deposition densities spanning more than three orders of magnitude from ~10^{12} atoms per m^2 in Ostrovskie (Vitebsk Oblast) to a maximum of 6.88×10^{15} in Chudiany in the rain-out spot in the Mogilev Oblast.

As with ^{129}I deposition, the data for ^{137}Cs also show that the intra-settlement differences tend to be only a factor of about two, i.e., 1.78±1.05.

Deposition of ^{131}I inferred from ^{129}I

The deposition densities of ^{131}I at the time of the accident inferred from our measured ^{129}I results are listed in Table 4. Also, compared in Figure 4 are the ratios est. ^{131}I-to-^{131}I and ^{129}I-to-^{137}Cs for each sampling site. In this case, "est. I-131" is the deposition of ^{131}I estimated to have occurred on April 26-28, 1986, based on our

Table 2. Deposition densities of I-129 in Belarus

Settlement & Sampling Site	Map #	I-129/m2	Settlement & Sampling Site	Map #	I-129/m2
Tulgovitchi-1	1	3.72E+14	Grandichi-1	39	3.75E+13
Tulgovitchi-2	2	1.20E+14	Grandichi-2	40	2.46E+13
Deriazshna-1	3	4.32E+14	Lunno-1	41	2.23E+13
Deryashnya-2	4	2.49E+14	Lunno-2	42	2.01E+13
Deryashnya-3	5	3.88E+14	Pogonnoe-1	43	8.27E+14
Veprin-1	6	1.45E+14	Pogonnoe-2	44	3.79E+14
Veprin-2	7	2.07E+14	Izabelino-1	45	2.71E+13
Gorodische-1	8	7.61E+12	Izabelino-2	46	1.86E+13
Gorodische-2	9	2.15E+13	Shevernichi-1	47	7.41E+12
Boloto-1	10	8.56E+12	Shevernichi-1	48	1.02E+13
Boloto-2	11	2.43E+13	Osipovichi-1	49	2.12E+13
Pruzhany-1	12	1.21E+13	Osipovichi-2	50	1.23E+13
Pruzhany-2	13	4.94E+12	N. Borschevka-1	51	3.57E+14
Ostrogliady-1	14	1.56E+14	N. Borschevka-2	52	5.76E+13
Ostrogliady-2	15	1.63E+14	Chudiany-1	53	3.05E+14
Bogino-1	16	1.32E+13	Rotki-1	54	3.88E+13
Bogino-2	17	2.30E+13	Bobr-1	55	1.50E+13
Obrub-1	18	9.36E+12	Bobr-2	56	3.18E+13
Obrub-2	19	2.93E+13	Pleschenitsy-1	57	1.01E+13
Holochie-1	20	1.53E+14	Pleschenitsy-2	58	2.26E+13
Holochie-2	21	3.63E+13	Losha-1	59	1.67E+13
Yankovichi-1	22	8.89E+12	Losha-2	60	2.54E+13
Yankovichi-2	23	2.28E+13	Zabolotye-1	61	2.10E+13
Lyntupy-1	24	1.75E+13	Zabolotye-2	62	1.68E+13
Lyntupy-2	25	2.43E+13	Krynka-1	63	1.54E+13
Babinichi-1	26	1.14E+13	Krynka-2	64	8.71E+12
Babinichi-2	27	2.07E+13	Kulikovka-1	65	1.07E+14
Korma-2	28	6.23E+13	Kulikovka-2	66	8.93E+13
Korma-1	29	5.19E+13	Bokhany-1	67	1.88E+13
Stodolichi-1	30	5.19E+13	Bokhany-2	68	1.44E+13
Stodolichi-2	31	5.39E+13	Volkovichi-1	69	3.17E+13
Simonichi-1	32	2.79E+13	Volkovichi-2	70	7.70E+13
Simonichi-2	33	2.86E+13	Veprin-1	71	1.96E+14
Yanovka-1	34	6.21E+13	Veprin-2	72	1.41E+14
Yanovka-2	35	3.79E+13	Bentyai-1	73	1.00E+13
Porechye-1	36	2.63E+13	Bentyai-2	74	1.36E+13
Porechye-2	37	1.63E+13	Ostrovskie-1	75	nd
Holochie-1	38	7.81E+13	Ostrovskie-2	76	nd

nd = not detectable above background level.

Table 3. Deposition densities of Cs-137 in Belarus

Settlement & Sampling Site	Map #	Cs-137/m2	Settlement & Sampling Site	Map #	Cs-137/m2
Tulgovitchi-1	1	7.76E+14	Grandichi-1	39	4.46E+12
Tulgovitchi-2	2	1.13E+15	Grandichi-2	40	5.68E+12
Deriazshna-1	3	3.15E+15	Lunno-1	41	3.02E+12
Deryashnya-2	4	5.08E+15	Lunno-2	42	6.62E+12
Deryashnya-3	5	4.29E+15	Pogonnoe-1	43	2.71E+15
Veprin-1	6	2.50E+15	Pogonnoe-2	44	3.65E+15
Veprin-2	7	2.61E+15	Izabelino-1	45	6.49E+12
Gorodische-1	8	3.74E+12	Izabelino-2	46	2.06E+12
Gorodische-2	9	2.61E+12	Shevernichi-1	47	7.14E+13
Boloto-1	10	1.55E+12	Shevernichi-1	48	8.16E+13
Boloto-2	11	6.12E+12	Osipovichi-1	49	4.20E+13
Pruzhany-1	12	5.65E+12	Osipovichi-2	50	9.20E+13
Pruzhany-2	13	7.50E+12	N. Borschevka-1	51	6.55E+13
Ostrogliady-1	14	1.63E+15	N. Borschevka-2	52	1.21E+14
Ostrogliady-2	15	6.13E+14	Chudiany-1	53	6.88E+15
Bogino-1	16	1.14E+12	Rotki-1	54	5.85E+14
Bogino-2	17	2.17E+12	Bobr-1	55	2.11E+13
Obrub-1	18	1.83E+12	Bobr-2	56	1.92E+13
Obrub-2	19	2.06E+12	Pleschenitsy-1	57	3.75E+12
Holochie-1	20	1.51E+15	Pleschenitsy-2	58	3.16E+12
Holochie-2	21	2.42E+14	Losha-1	59	6.65E+12
Yankovichi-1	22	2.68E+12	Losha-2	60	1.02E+13
Yankovichi-2	23	2.86E+12	Zabolotye-1	61	1.37E+13
Lyntupy-1	24	3.31E+12	Zabolotye-2	62	1.01E+13
Lyntupy-2	25	2.06E+12	Krynka-1	63	9.38E+12
Babinichi-1	26	6.22E+12	Krynka-2	64	5.08E+12
Babinichi-2	27	2.06E+12	Kulikovka-1	65	1.51E+15
Korma-2	28	7.28E+14	Kulikovka-2	66	1.49E+15
Korma-1	29	5.49E+14	Bokhany-1	67	1.36E+13
Stodolichi-1	30	1.32E+14	Bokhany-2	68	1.39E+13
Stodolichi-2	31	7.40E+13	Volkovichi-1	69	1.48E+14
Simonichi-1	32	8.10E+13	Volkovichi-2	70	3.28E+14
Simonichi-2	33	7.48E+13	Veprin-1	71	1.52E+15
Yanovka-1	34	1.39E+14	Veprin-2	72	1.36E+15
Yanovka-2	35	1.74E+14	Bentyai-1	73	2.60E+12
Porechye-1	36	1.78E+13	Bentyai-2	74	nd
Porechye-2	37	1.25E+13	Ostrovskie-1	75	nd
Holochie-1	38	6.64E+14	Ostrovskie-2	76	nd

Table 4. Deposition densities of I-131 inferred from I-129 in Belarus

Settlement & Sampling Site	Map #	est. I-131/m2	Settlement & Sampling Site	Map #	est. I-131/m2
Tulgovitchi-1	1	2.48E+13	Grandichi-1	39	2.50E+12
Tulgovitchi-2	2	7.99E+12	Grandichi-2	40	1.64E+12
Deriazshna-1	3	2.88E+13	Lunno-1	41	1.49E+12
Deryashnya-2	4	1.66E+13	Lunno-2	42	1.34E+12
Deryashnya-3	5	2.59E+13	Pogonnoe-1	43	5.51E+13
Veprin-1	6	9.67E+12	Pogonnoe-2	44	2.53E+13
Veprin-2	7	1.38E+13	Izabelino-1	45	1.81E+12
Gorodische-1	8	5.07E+11	Izabelino-2	46	1.24E+12
Gorodische-2	9	1.43E+12	Shevernichi-1	47	4.94E+11
Boloto-1	10	5.71E+11	Shevernichi-1	48	6.79E+11
Boloto-2	11	1.62E+12	Osipovichi-1	49	1.41E+12
Pruzhany-1	12	8.09E+11	Osipovichi-2	50	8.22E+11
Pruzhany-2	13	3.29E+11	N. Borschevka-1	51	2.38E+13
Ostrogliady-1	14	1.04E+13	N. Borschevka-2	52	3.84E+12
Ostrogliady-2	15	1.09E+13	Chudiany-1	53	2.03E+13
Bogino-1	16	8.80E+11	Rotki-1	54	2.59E+12
Bogino-2	17	1.53E+12	Bobr-1	55	1.00E+12
Obrub-1	18	6.24E+11	Bobr-2	56	2.12E+12
Obrub-2	19	1.95E+12	Pleschenitsy-1	57	6.71E+11
Holochie-1	20	1.02E+13	Pleschenitsy-2	58	1.51E+12
Holochie-2	21	2.42E+12	Losha-1	59	1.11E+12
Yankovichi-1	22	5.93E+11	Losha-2	60	1.69E+12
Yankovichi-2	23	1.52E+12	Zabolotye-1	61	1.40E+12
Lyntupy-1	24	1.17E+12	Zabolotye-2	62	1.12E+12
Lyntupy-2	25	1.62E+12	Krynka-1	63	1.02E+12
Babinichi-1	26	7.63E+11	Krynka-2	64	5.81E+11
Babinichi-2	27	1.38E+12	Kulikovka-1	65	7.13E+12
Korma-2	28	4.15E+12	Kulikovka-2	66	5.95E+12
Korma-1	29	3.46E+12	Bokhany-1	67	1.25E+12
Stodolichi-1	30	3.46E+12	Bokhany-2	68	9.58E+11
Stodolichi-2	31	3.59E+12	Volkovichi-1	69	2.11E+12
Simonichi-1	32	1.86E+12	Volkovichi-2	70	5.13E+12
Simonichi-2	33	1.91E+12	Veprin-1	71	1.31E+13
Yanovka-1	34	4.14E+12	Veprin-2	72	9.43E+12
Yanovka-2	35	2.53E+12	Bentyai-1	73	6.67E+11
Porechye-1	36	1.75E+12	Bentyai-2	74	9.73E+12
Porechye-2	37	1.08E+12	Ostrovskie-1	75	nd
Holochie-1	38	5.20E+12	Ostrovskie-2	76	nd

contemporary ^{129}I deposition measurements. "I-131" is the deposition density of ^{131}I actually measured soon after the accident (and back-decayed to April 26-28, 1986) in soil samples obtained from some (not all) of the settlements that we have sampled. "I-129" and "Cs-137" are the deposition densities of these isotopes that we have measured in Belarus (refer to sampling sites in Table 1 and Figure 1).

Importantly, it is clear from Figure 4 that the correlation between ^{131}I inferred from our measured ^{129}I and ^{131}I actually measured in the same settlements immediately after the accident in 1986 is much better than the correlation between ^{129}I and ^{137}Cs, even though ^{129}I and ^{137}Cs were measured in the same core samples. This demonstrates the expected result that ^{129}I is superior to ^{137}Cs as a surrogate for ^{131}I. It is noted that ^{137}Cs is presently being used as a surrogate for ^{131}I to reconstruct thyroid doses in regions with little or no direct thyroid measurements. Clearly, ^{129}I should be used as a surrogate instead of ^{137}Cs in such regions.

Total iodine concentrations in Belarus soil

The total iodine concentrations measured in Belarus soil are listed in Table 5. These data are illustrated in Figure 5 and show the large number of measurements made in this project across the entire country of Belarus. Importantly, the iodine concentrations (mostly below 1 μg/g) are one to two orders of magnitude lower than the iodine concentrations in the Marshall Islands and Japan, places where other thyroid-cancer studies have been performed. Also, the Belarus soil concentrations are factors of five to ten lower than those measured in the western United States, which tend to be in the 5 μg/g range. For Belarus (and probably also for other countries in the region), the low iodine concentration in the local environment, and apparently in the diet generally, may be an issue that deserves further evaluation in connection with its potential influence on radiogenic risk for thyroid cancer.

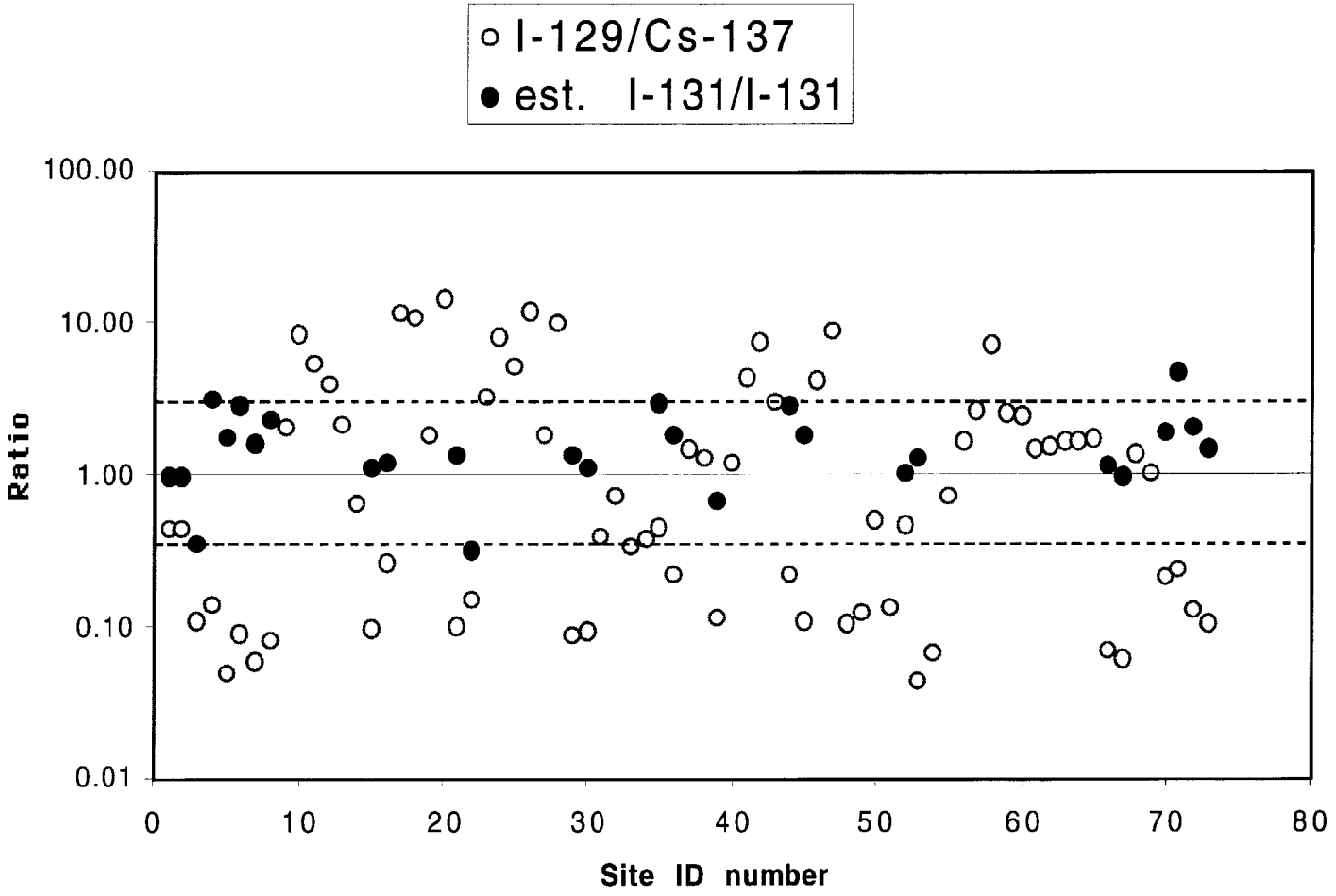

Figure 4. Comparison of ^{129}I and ^{137}Cs as surrogates for ^{131}I deposition in Belarus. The data plotted are the ratio $^{129}I/^{137}Cs$ (open circles) and the ratio est. ^{131}I/measured ^{131}I (solid circles), where est. ^{131}I was inferred from our ^{129}I measurements by dividing the ^{129}I deposition densities by 15 (see text). Note that about 90% of the solid circles are within the dashed lines while a large number of the open circles are outside the lines, demonstrating that ^{129}I is a better than ^{137}Cs as a surrogate for ^{131}I deposition.

Table 5. Total iodine concentrations in Belarus

Settlement & Sampling Site	Map #	Iodine µg/g	SD	Settlement & Sampling Site	Map #	Iodine µg/g	SD
Tulgovitchi-1	1	0.3155	0.0122	Grandichi-1	39	0.3520	0.0161
Tulgovitchi-2	2	0.2615	0.0503	Grandichi-2	40	2.4507	0.1426
Deriazshna-1	3	0.3543	0.0374	Lunno-1	41	0.5879	0.0316
Deryashnya-2	4	0.6458	0.0755	Lunno-2	42	1.0337	0.0334
Deryashnya-3	5	1.1904	0.0718	Pogonnoe-1	43	0.9120	0.0299
Veprin-1	6	1.1392	0.0296	Pogonnoe-2	44	0.9960	0.0259
Veprin-2	7	0.4885	0.0064	Izabelino-1	45	1.3121	0.0515
Gorodische-1	8	0.5907	0.0353	Izabelino-2	46	0.8366	0.0380
Gorodische-2	9	2.2898	0.1260	Shevernichi-1	47	0.4022	0.0258
Boloto-1	10	0.3883	0.0286	Shevernichi-2	48	0.4939	0.0161
Boloto-2	11	0.6427	0.0848	Osipovichi-1	49	0.7202	0.0226
Pruzhany-1	12	0.5644	0.0080	Osipovichi-2	50	0.3529	0.0099
Pruzhany-2	13	0.1453	0.0028	N. Borschevka-1	51	0.8452	0.0219
Ostrogliady-1	14	0.9426	0.1131	N. Borschevka-2	52	1.6185	0.0741
Ostrogliady-2	15	0.5001	0.0131	Chudiany-1	53	1.0246	0.0475
Bogino-1	16	0.8233	0.0342	Rotki-1	54	0.5331	0.0160
Bogino-2	17	0.6380	0.0406	Bobr-1	55	0.8583	0.0347
Obrub-1	18	0.2678	0.0344	Bobr-2	56	0.6544	0.0344
Obrub-2	19	1.1184	0.0664	Pleschenitsy-1	57	1.7440	0.0543
Holochie-1	20	1.6200	0.1055	Pleschenitsy-2	58	0.9389	0.0561
Holochie-2	21	2.8846	0.0761	Losha-1	59	0.8573	0.0320
Yankovichi-1	22	0.2590	0.0087	Losha-2	60	0.9469	0.1094
Yankovichi-2	23	0.8723	0.0274	Zabolotye-1	61	1.2044	0.0408
Lyntupy-1	24	0.8488	0.0175	Zabolotye-2	62	1.3644	0.0460
Lyntupy-2	25	0.6697	0.0450	Krynka-1	63	0.8596	0.0735
Babinichi-1	26	0.7872	0.0275	Krynka-2	64	0.7825	0.0633
Babinichi-2	27	0.7802	0.0420	Kulikovka-1	65	0.7114	0.0275
Korma-2	28	1.7639	0.0706	Kulikovka-2	66	0.8819	0.0397
Korma-1	29	0.4599	0.0296	Bokhany-1	67	2.3126	0.1645
Stodolichi-1	30	0.5527	0.0239	Bokhany-2	68	0.7235	0.0484
Stodolichi-2	31	0.3645	0.0247	Volkovichi-1	69	1.7626	0.1681
Simonichi-1	32	0.8042	0.0683	Volkovichi-2	70	1.6869	0.1621
Simonichi-2	33	0.6104	0.0408	Veprin-1	71	1.0226	0.0783
Yanovka-1	34	0.4778	0.0579	Veprin-2	72	0.7217	0.0304
Yanovka-2	35	0.7092	0.0185	Bentyai-1	73	0.6431	0.0340
Porechye-1	36	0.7679	0.0659	Bentyai-2	74	2.3499	0.1073
Porechye-2	37	0.5546	0.0321	Ostrovskie-1	75	0.7786	0.0186
Holochie-1	38	0.9715	0.0411	Ostrovskie-2	76	1.3628	0.0562

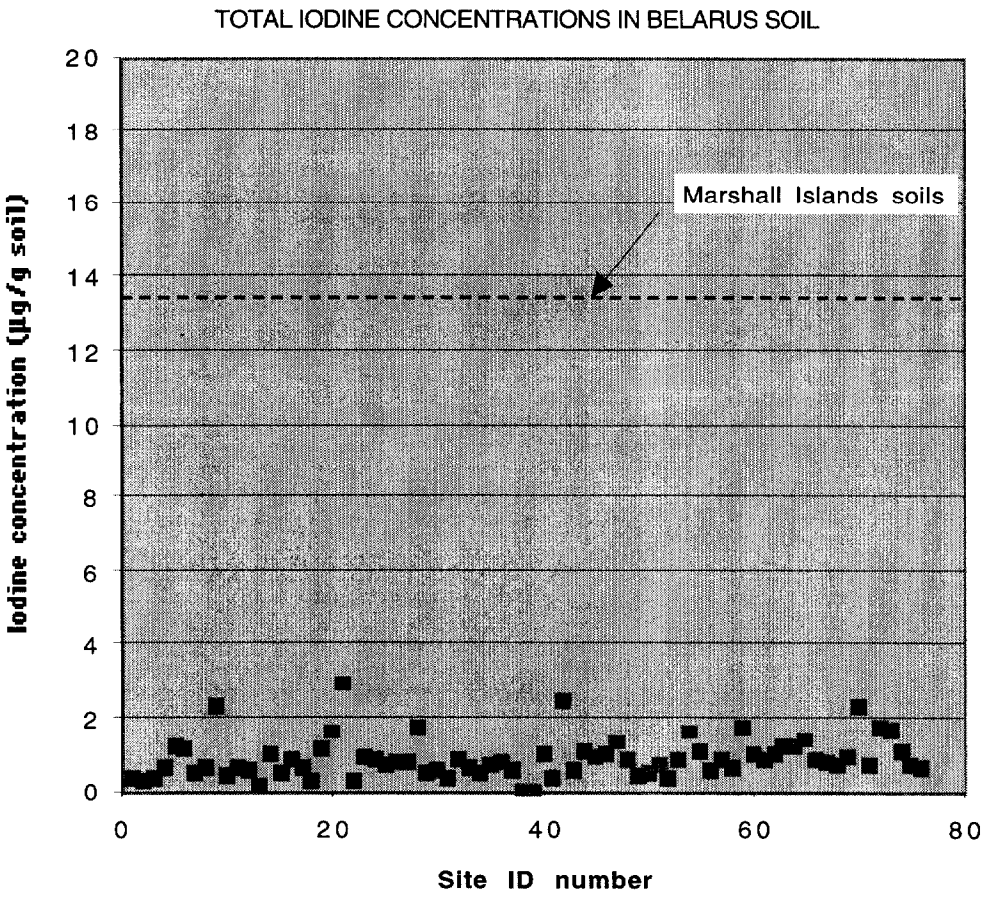

Figure 5. Total iodine concentrations in Belarus soil. The soil concentration in the Marshall Islands is included for comparison.

CONCLUSIONS CONCERNING THE SCIENTIFIC/TECHNICAL DATA

The following conclusions can be drawn from the available data:

- Both ^{129}I and ^{137}Cs are retained firmly in the top ~15 to 20 cm of the soil and thus the possibility for obtaining samples for deposition-density measurements will remain.

- Deposition densities of ^{129}I can be measured accurately using the methods and AMS capabilities developed under the Feasibility Study (e.g., Straume et al. 1996, Marchetti et al. 1997) and the present Follow-on Study.

- The results show good agreement between ^{131}I deposition inferred from our ^{129}I measurements and ^{131}I actually measured in the same settlements in 1986.

- In contrast, the correlation between ^{129}I and ^{137}Cs deposition is poor, hence ^{137}Cs is not a reliable surrogate for ^{131}I.

- Indications are that total iodine concentrations in top soil from Belarus are low compared with other regions of the world where radiogenic thyroid cancer has been studied (e.g., Japan and Marshall Islands, where iodine is obtained from sea food diets as well). This suggests the need for additional evaluations to explore whether the relatively low iodine concentrations in Belarus soil resulted in low concentrations in the diet and whether this might have modified the risk of developing radiogenic thyroid cancer.

- Finally, the use of ^{129}I deposition data measured by AMS appears to be the only method currently available to obtain reliable radioiodine deposition densities in regions where direct iodine measurements were absent or inadequate and, also, to provide confirmation and QC in areas where such measurements were made.

MILESTONES AND DELIVERABLES

I. Milestones & Status of Completion

1. Develop a joint agreement between study participants in Belarus, Germany, and the US.

   Completed in May 1997.

2. Develop a technical protocol for obtaining and processing soil samples.

   Completed in May 1997.

3. Jointly with the Research and Clinical Institute of Radiation Medicine and Endocrinology (Minsk), GSF (Munich), and the University of Utah, provide support and perform a soil sampling expedition that includes settlements throughout the country of Belarus.

   Completed in July 1997 (see Table 1 and Figure 1 for sampling locations).

4. Provide training for a Minsk chemist at the University of Utah in procedures for iodine extraction and measurement.

   Completed in October 1997.

5. Procurement and shipment of laboratory equipment required to set-up the Minsk iodine laboratory.

   Completed in February 1998.

6. Shipment of soil samples from Minsk to the University of Utah.

   Completed in June 1998.

7. Extraction of iodine and measurement of ^{129}I, total iodine, and ^{137}Cs in soil samples. Determination of deposition densities for ^{129}I, ^{131}I, and ^{137}Cs from our measurements together with the ratio for ^{129}I-to-^{131}I at the time of the accident.

   Completed in September 1999.

II. Other Relevant Information

• As promised, this project was successfully completed in FY99 with FY98 funding without the need for additional support from DOE.

• It was originally planned that a sub-set of the samples collected in Belarus would be measured for ^{129}I by GSF in Munich. However, they informed us that they lost their capability to measure ^{129}I and therefore all measurements were made by Dr. Straume in Utah. This added some unexpected cost to the effort which was absorbed by the University of Utah.

III. Future Use of this Capability

It is now fully expected that this unique capability will be used to improve the thyroid dosimetry in Belarus, Ukraine, and parts of Russia contaminated by Chernobyl fallout. NCI is interested in using ^{129}I to help in their thyroid dosimetry efforts in support of Chernobyl health studies. During the past year, Prof. Straume has been working with NCI to help define specific applications.

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